Climbing the Jaynes–Cummings ladder by photon counting

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We present a new method to observe direct experimental evidence of Jaynes–Cummings nonlinearities in a strongly dissipative cavity quantum electrodynamics system, where large losses compete with the strong light-matter interaction. This is a highly topical problem, particularly for quantum dots in microcavities where transitions from higher rungs of the Jaynes–Cummings ladder remain to be evidenced explicitly. We compare coherent and incoherent excitations of the system and find that resonant excitation of the detuned emitter make it possible to unambiguously evidence few photon quantum nonlinearities in currently available experimental systems.

After reaching the strong coupling regime of the light-matter interaction for optically active quantum dots (QDs) in high quality microcavities [1–3] and demonstrating its single-photon character [4, 5], a major remaining challenge is to obtain clear and direct evidence of quantum nonlinearities. Indirect manifestations have already been provided in the form of photon blockade [6–8] or broadening of the Rabi doublet due to excited states [9] but these are unspecific with regard to their origin and whether such quantum effects are described by the Jaynes–Cummings (JC) Hamiltonian,

$$H = \omega_a a^\dagger a + (\omega_a - \Delta)\sigma^\dagger\sigma + g(a^\dagger\sigma + a\sigma^\dagger)\,, \qquad (1)$$

the paradigm of quantum interaction between quanta of light (with Bose operator a) and a two-level system (σ). Such quantum nonlinearities have been demonstrated in cavity QED systems, such as atoms [10, 11] or, perhaps most spectacularly, for superconducting qubits [12, 13], where the fingerprints of JC physics have been observed, in particular the anharmonic splitting of states with the number of excitation [14]. Only very recently have semiconductor systems begun to exhibit this rich phenomenology [15]. The difficulty of directly observing transitions between the different rungs of the JC ladder in semiconductors can, presumably, be traced to the strong dephasing in these systems. Indeed in the spectral domain the uncertainty due to the short photon lifetime washes out completely the weak square root dependence of the splitting of the excited states. Any traces of the quantum interaction, lost in the energy of the emitted photons, is however recovered in the statistics of the emitted photons [16].

In this Letter, we highlight that, although it is difficult to obtain clear evidence of the JC ladder in state-of-the-art semiconductor samples when detecting luminescence under incoherent excitation, it is however possible to observe clear signatures of the higher rungs by performing photon counting measurements to probe the statistics of the emitted photons from the cavity. We show that this process is optimum when coherently exciting the detuned QD, which result in strong photon bunching at the resonances of the JC ladder. Our results provide a route for experimentalists to test the suitability of JC model to describe QD-cavity systems, which have displayed many variations from their counterparts in atomic or superconducting cavity QED [17, 18].

The effect of dissipation can be introduced into the JC Hamiltonian with Liouville equation $\partial_t\rho = \mathcal{L}(\rho)$, where the so-called Liouvillian $\mathcal{L}$ adds a non-unitary evolution of the density matrix ρ to the Hamiltonian dynamics:

$$\mathcal{L}(\rho) = i[\rho, H] + \frac{\gamma_a}{2}\mathcal{L}_a(\rho) + \frac{\gamma_\sigma}{2}\mathcal{L}_\sigma(\rho) + \frac{P_a}{2}\mathcal{L}_{a^\dagger}(\rho) + \frac{P_\sigma}{2}\mathcal{L}_{\sigma^\dagger}(\rho)\,, \qquad (2)$$

where $\mathcal{L}_c(\rho) = 2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c$. This describes the decay (at rate γ_a for the cavity photon and γ_σ for the QD exciton) or excitation (P_a and P_σ) due to incoher-

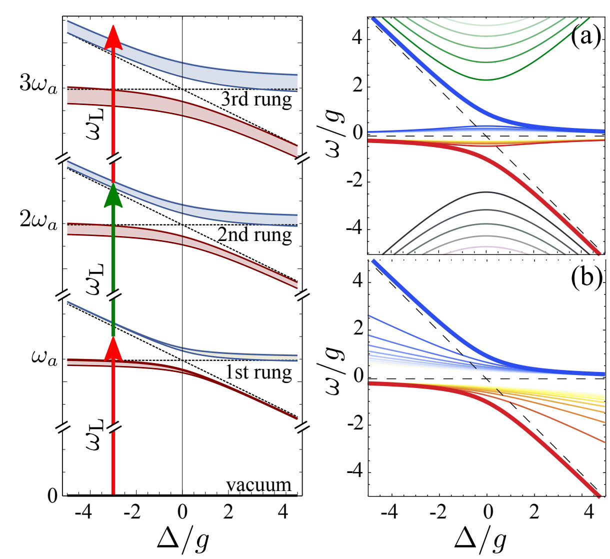

FIG. 1. (color online) Energies and linewidths of the first three rungs of the dissipative JC ladder ($\gamma_a = g$) as function of detuning. The configuration of the two-photon blockade is indicated by the arrows. Transitions energies and resonances of the JC ladder as a function of detuning, probed under incoherent (a) and coherent (b) excitation, respectively. The thick solid lines are the upper and lower polariton lines of the first rung. The thin dotted lines are the bare (undressed) QD and cavity. They sandwich *inner lines* and are encompassed by *outer lines* from transitions higher in the ladder.

ent pumping [19]. Dephasing can be included in this formalism with additional terms $\mathcal{L}_{\sigma^\dagger\sigma}$ for pure dephasing [3, 20, 21] or $\mathcal{L}_{\sigma^\dagger a}$ for phonon induced dephasing [22]. We have checked that unless these quantities have very large values, they do not affect qualitatively our findings.

As a result of finite lifetime, the energies of the JC system become complex. They are obtained by diagonalizing the Liouvillian (2):

$$E_\pm^k = k\omega_a - \frac{\Delta}{2} - i\frac{(2k-1)\gamma_a + \gamma_\sigma}{4} \pm \sqrt{(\sqrt{k}g)^2 - \left(\frac{\gamma_a - \gamma_\sigma}{4} + i\frac{\Delta}{2}\right)^2}\,, \quad (3)$$

where $E_\pm^k$ corresponds to the kth rung of the system. This is a generalization of the usual expression that neglects lifetime. It shows that detuning behaves as an effective dissipation and, thus, strong coupling is optimum at resonance. Following Eq. (3) we plot the eigenenergies of a dissipative JC system ($\gamma_a = g$) in Fig. 1 as function of detuning. Here, the broadening $\pm 2\Im(E_\pm)$ is visualized as the width of the line. The first rung ($k = 1$) is the familiar anticrossing of two coupled modes, that describes equally well the linear quantum regime and a classical system [23]. Higher rungs ($k > 1$) reproduce the same pattern with two variations with respect to the linear case (at resonance): the splitting increases as $\sqrt{k}$ and the broadedning as $k\gamma_a/g$. Since the increase of the coupling rate with the number of excitations (k) is slower than the decoherence, climbing the ladder makes it increasingly difficult to observe the quantum features. One could circumvent this problem by decreasing γ_a, but in typical semiconductor systems $\gamma_a \approx g \gg \gamma_\sigma$, which is the configuration we will focus on in the following (we shall in fact assume $\gamma_\sigma = 0$). However, it is advantageous if γ_a is not too small: it increases signal intensity and better preserves the statistics of the state prepared inside the cavity. These two qualities are essential for working quantum devices. Experimentally, the energy structure of the ladder cannot be observed directly and the way it manifests itself depends on the kind of measurement performed and how the system is excited. There are countless variations of experiments, but most can be categorized in *incoherent* and *coherent* excitation. In the former case, one populates the energy levels of the system through relaxation of charge carriers, and observes the emission at energies corresponding to transitions between consecutive manifolds. In the latter case, a well defined energy is incident on the system and one observes its direct response via a number of observables. The expected resonances of the system in these two configurations are displayed in Figs. 1(a-b), respectively. The actual observation is a combination of these lines, depending on the interplay of their oscillator strengths and the fluctuations in population. Both have in common the upper polariton (UP) and lower polariton (LP) lines (thick lines) of the first rung, which are most easily excited and detected.

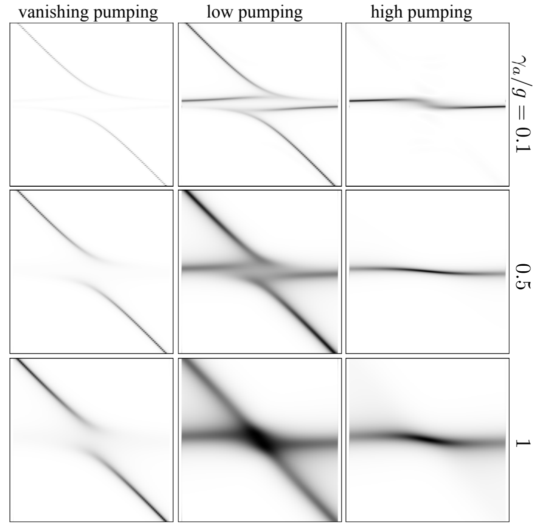

FIG. 2. Incoherent excitation: cavity photoluminescence spectra for increasingly dissipative systems from upper to lower row ($\gamma_a/g = 0.1$, 0.5 and 1), and for increasing excitation power from left to right column. Quantum nonlinearities are more clearly observed for a small, but nonvanishing incoherent excitation. Higher pumping brings the system into lasing or collapses the Rabi doublet. Only in very-strongly coupled systems does the photoluminescence reconstruct the JC ladder, albeit with the outer transitions being much suppressed in the cavity emission. Axes are not shown for clarity but are the same as Fig. 1(a).

To prove the quantum character of this system, the field quantization needs to be demonstrated, and this requires observing at least some of the lines that arise from higher rungs of the ladder.

In the incoherent excitation case [Fig. 1(a)], there are two sets of additional lines, one in-between the Rabi doublet, the other sandwiching it. The *inner lines* are narrowly packed together and are broader, since they arise from higher rungs, thus demanding an extraordinarily good system to resolve them. The *outer lines* have a much higher splitting between them. However, the strength of these transitions is strongly suppressed in the cavity emission, since the emitter is de-excited at the same time as the photon is emitted, whilst the cavity favours the sole emission of a photon. In Fig. 2, we show the cavity photoluminescence spectra $\langle a^\dagger(\omega)a(\omega)\rangle$ [24] as function of detuning for increasing dissipation from top to bottom and for various intensities of incoherent excitation from left to right. The quality of the strong coupling ranges from significantly better than is currently available ($\gamma_a/g = 0.1$), via state-of-the-art systems ($\gamma_a/g = 0.5$ [25]) to the typical value available in many laboratories worldwide ($\gamma_a/g = 1$ [20]). These density plots show how the JC structure of Fig. 1(a) manifests itself in photoluminescence. In all cases the outer lines are indeed suppressed. The main features are the upper and lower polaritons. When one tries to climb the ladder

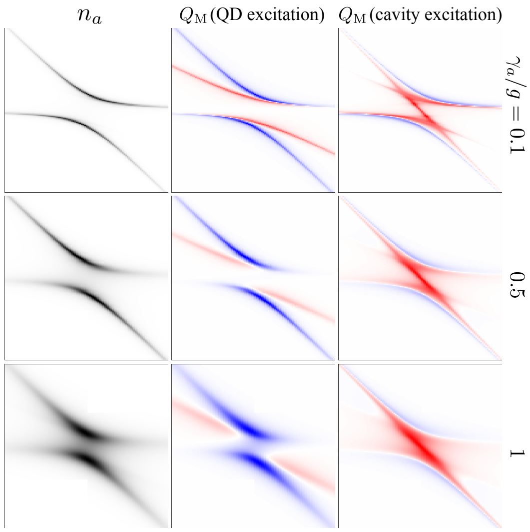

FIG. 3. (color online) Coherent excitation: cavity intensity n_a (first column), and Mandel factor $Q_{\rm M}$ under coherent QD (second column) and cavity (third column) excitation, for increasingly dissipative systems. Blue and red refer to negative (antibunching) and positive (bunching) values, respectively. While intensity (or other observables such as absorption or scattering) elicit a response only from the bottom of the ladder, the photon statistics displays strong features from the 2nd rung when exciting the QD detuned from the cavity. The signature is then unambiguous even for very dissipative systems. For cavity excitation, bare modes dominate. Axes are not shown for clarity but are the same as Fig. 1(b).

by increasing pumping, only in the very best system can additional lines of the second rung be clearly resolved. In the case of $\gamma_a/g = 0.5$, although a strong deviation from the anticrossing is observed, no clear fingerprints of the JC features are observed. At resonance, only a doublet is observed (qualitatively similar to the Rabi doublet) and out of resonance, a triplet is observed [26]. This might in fact be consistent with the experimental situation of Ref. [17], that did not make any claim in this direction. For smaller strong-coupling, although still deviating from crossing or anticrossing, there is again no useful characterisation of the JC physics. At pumping levels higher than those presented in Fig. 2, the system moves into the lasing regime [25] and the JC description is not adequate anymore [27].

In the coherent excitation case, Fig. 1(b), there are only inner lines, but they are clearly separated from one another in the energy-detuning space. These arise from multiple-photon excitations. When the laser is at an energy $\omega_{\rm L}$, smaller than the upper polariton $\omega_{\rm UP}$ energy, as indicated by the arrows in Fig. 1, it cannot excite the system with one photon. However, if $2\omega = \Re(E_+^2)$, then it can access the second rung by a two-photon excitation process. At the point highlighted, the laser is blocked at the first rung, is resonant with the second rung, and is blocked again at the third rung, even when taking into account the large broadening of higher excited manifolds. This configuration, therefore, efficiently filters out the two-photon fluctuation of the laser and performs a type of *two-photon blockade*, in analogy with the photon blockade effect [28, 29], where blocking from the second rung is used to produce a single-photon source [6, 7]. This scheme does not work so well at resonance because of overlap of the transitions broadened by dissipation.

Furthermore, as for incoherent excitation, one should try to avoid overly strong pumping for coherent excitation. In the Hamiltonian it is included by adding the term $\Omega_a \exp(i\omega t) a^\dagger$ + h.c. for coherent cavity excitation [6–8]. One can also excite the QD coherently $\Omega_\sigma \exp(i\omega t)\sigma^\dagger$ + h.c., e.g., by side emission in a pillar microcavity. At low driving intensity, one only sees clearly the lower and upper polariton of the first rung, but for increasing pumping the coherent excitation quickly dresses the states and distorts Eq. (3). In the first column of Fig. 3 we plot the cavity population $n_a = \langle a^\dagger a\rangle$ when driving the system coherently for increasingly dissipative systems from top to bottom. The plots fail to reproduce the nonlinear features of Fig. 1(b), indicating that the intensity ($\propto n_a$)—and in fact other observables involving first order correlators such as reflectivity, transmission, absorption, etc.—are not optimal to resolve the higher rungs of the JC ladder. In all these cases, there is a strong response when the laser hits the polariton resonances of the first rung, but otherwise the response is weak. There is, however, a strong response in observables involving higher order correlators (at zero time delay), of the type $G^{(n)} = \langle a^{\dagger n} a^n\rangle$, which are linked to photon counting. One usually deals with the normalized quantities, $g^{(n)} = G^{(n)}/n_a^n$. In particular, $g^{(2)}$ is popular as the standard to classify antibunched (non-classical) ($g^{(2)} = 0$), Poissonian (coherent) ($g^{(2)} = 1$) and bunched (chaotic/thermal) ($g^{(2)} = 2$) light sources. Other related quantities are more useful in this context [30], such as the Mandel parameter $Q_{\rm M}$:

$$Q_{\rm M} = (g^{(2)} - 1) n_a \tag{4}$$

which sign provides the (anti)bunched character of the statistics, while also taking into account the available signal. The $Q_{\rm M}$ parameter of the light emitted by the cavity is shown in Fig. 3 for coherent excitation of the QD (middle column) and of the cavity (right column). Remarkably, the middle column shows that this measurement unravels the second rung of the JC ladder: when the coherent excitation source is tuned to the 1st or the 2nd resonance, the photon statistics sharply responds. As detailed in Fig. 4(a-b), the statistics changes its character from antibunching on the polariton to bunching on the 2nd rung, caused by the two-photon blockade configuration displayed in Fig. 1. As a result of the separation of the resonances, one can clearly observe both resonances in this measurement with QD excitation. The scheme is much less efficient when exciting the cavity, with a

strong response on the bare QD, as can be seen both on the density plot in Fig. 3 (right column) or on the cut in Fig. 4(a-b). This is due to a sudden drop of the intensity when exciting the cavity at the energy of the QD.

These results show that even in very dissipative systems, where the coupling rate is of the order of the decay rate, the 2nd rung can be unambiguously observed in experiment, by combining detuning, QD excitation and photon counting. Remarkably, this scheme can be extended to higher rungs of the ladder, still with systems that operate in the presence of significant dissipation. Measuring the differential correlation function:

$$C^{(n)} = \langle a^{\dagger n} a^n \rangle - \langle a^\dagger a \rangle^n \,, \tag{5}$$

which quantifies the deviation of coincidences from uncorrelated, Poissonian events, one finds sharp resonances at the nth photon resonance condition. In Fig. 4(c) we plot $C^{(n)}$ up to the fourth rung, still for coherent excitation of the dot, when it is detuned by $\Delta/g = 4$ from the cavity. Such high-order coincidences can be measured by recently developed experimental techniques such as photon counting using a streak camera [31]. The signal is increasingly difficult to obtain for higher order as it requires to accumulate statistics for increasingly unlikely events (curves have been rescaled as indicated in the figure). However, given a sufficiently strong signal, one obtains sharp resonances precisely located at the JC multi-photon resonances, even for very dissipative systems such as those currently available. The scheme is robust to increased pumping, which broadens the resonances but does not appreciably shift their maxima, and provides more signal. Although the experiment to perform is the same, the quantity to analyse is $C^{(n)}$ rather than $g^{(n)}$ which is shown in Fig. 4(d). One sees that $g^{(n)}$ increases with n, which marks the higher quantumness of light emitted when hitting the higher rungs. However, the signal also becomes exponentially dimmer as a result. Because of fluctuations to all orders, the resonances are also not exactly mapped with the transitions [compare the agreement of $g^{(2)}$ with the theoretical line at low pumping in (a) and its disagreement at high pumping in (d)], in contrast to $C^{(n)}$ that follow them accurately (except very close to resonance).

In conclusion, we have proposed an experimental scheme able to unravel the JC nonlinearities in dissipative QD-cavity systems similar to those currently available. We show that by analysing the photon counting statistics of the light emitted by a cavity when the quantum emitter is detuned and excited coherently, one can observe a clear and unambiguous fingerprint of quantum nonlinearities. The demonstration of JC physics of QDs in microcavities, will pave the way for practical, working quantum information devices in the solid state.

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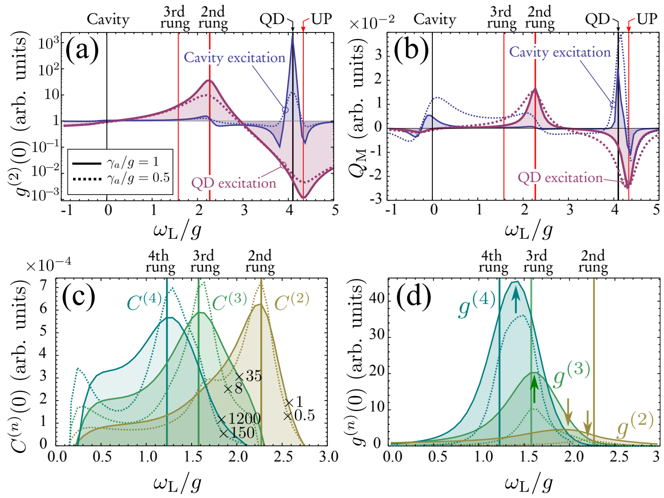

FIG. 4. (color online) Quantum statistics of the cavity photons under coherent excitation for a good (dashed lines) and a typical (solid lines) system. (a) and (b): $g^{(2)}$ and Q_{M} parameter, showing how the 2nd rung transition is more clearly identified under QD excitation. Q_{M} is preferable to $g^{(2)}$ as it takes into account the available signal. (c) Differential correlations $C^{(n)}$ that peak sharply at the nth resonance, thus clearly identifying the higher states of the ladder. (d) nth order correlation functions $g^{(n)}$, that are loosely connected to the theoretical transitions. Maxima are indicated by arrows.

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